

* NOTICES *

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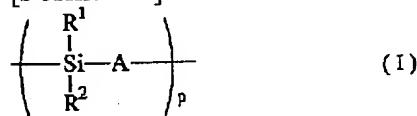
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CLAIMS

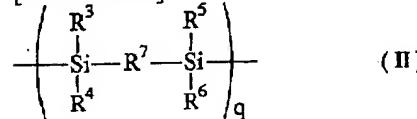
[Claim(s)]

[Claim 1] The silicon content copolymerization polymer which number average molecular weight is 500-1,000,000, and is characterized by including the structural unit expressed with the following general formula (I) and (II) at least.

[Formula 1]



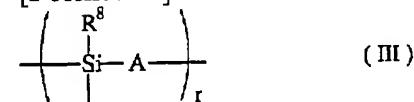
[Formula 2]



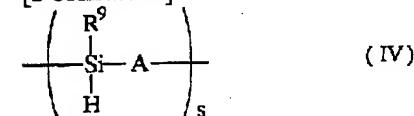
R1, R2, R3, R4, R5, and R6 are an alkyl group, an alkenyl radical, a cycloalkyl radical, an aryl group, an aralkyl radical, an alkylamino radical, an alkyl silyl radical, or an alkoxy group independently among [upper type, respectively, R7 is the radical of bivalence, and A is NH or O. Moreover, a structural unit (I) and (II) are random, each mole ratio p and q is $q/(p+q) = 0.01-0.99$, and the rate of Si-O association in a polymer and Si-N association is $Si-O/(Si-N+Si-O) = 0.01-0.99$.]

[Claim 2] The silicon content copolymerization polymer according to claim 1 to which a silicon content copolymerization polymer is further characterized by including a kind of structural unit of the following general formula (III) and (IV) at least.

[Formula 3]



[Formula 4]

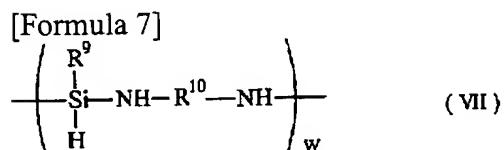
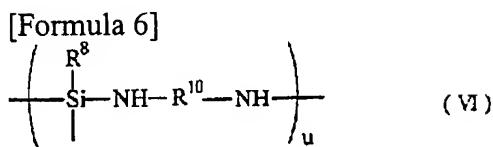
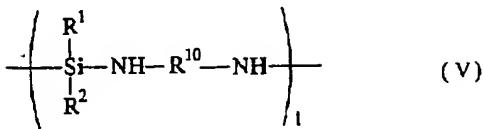


R8 and R9 are an alkyl group, an alkenyl radical, a cycloalkyl radical, an aryl group, an aralkyl radical, an alkylamino radical, an alkyl silyl radical, or an alkoxy group independently among [upper type, respectively, and A is NH or O. Moreover, the structural unit in a polymer (I) to (IV) is random, and each mole ratio p, q, r, and s takes the following relation.

$$q/(p+q+r+s) = 0.01-0.99$$

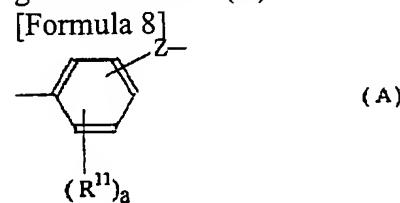
[Claim 3] The silicon content copolymerization polymer according to claim 1 or 2 to which a silicon content copolymerization polymer is further characterized by including at least a kind of structural unit to (VI) VI [the following general formula (V) and] Attain to (VII).

[Formula 5]

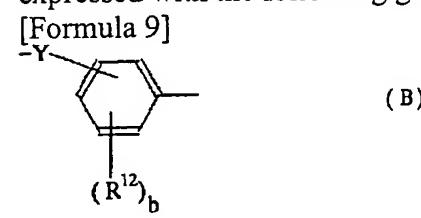


R1, R2, R8, and R9 are an alkyl group, an alkenyl radical, a cycloalkyl radical, an aryl group, an aralkyl radical, an alkylamino radical, an alkyl silyl radical, or an alkoxy group independently among [upper type, respectively, and R10 is the aromatic series radical of bivalence. Moreover, a structural unit (V) - (VII) are random.]

[Claim 4] The silicon content copolymerization polymer according to claim 1 to 3 characterized by the aromatic series radical of said bivalence being a radical expressed with an aralkylene group, a naphthylene radical, or the following general formula (A).

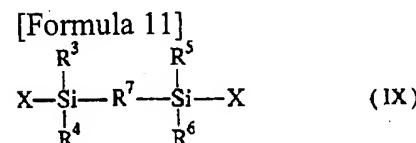
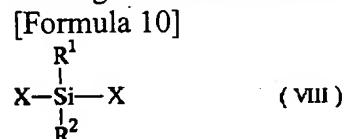


R11 is a radical which a halogen atom or a low-grade alkyl group, and a couple directly the integer of 0-4, and Z, or is expressed with the following general formula (B) among [upper type.



(or [that R12 couples directly the integer of 0-4, and Y for a halogen atom or a low-grade alkyl group, and b among a formula] -- or it is the radical of bivalence.)]

[Claim 5] The manufacture approach of the silicon content copolymerization polymer according to claim 1 characterized by making the mixture which contains at least the disilyl compound expressed with the ORGANO PORIHARO silane expressed with the following general formula (VIII), and the following general formula (IX) react with the water which the suitable solvent was made to distribute, carrying out a reaction with ammonia after that and making an unreacted halo silane react completely.

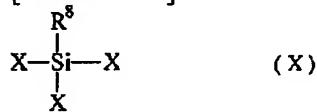


(R1, R2, R3, R4, R5, and R6 are an alkyl group, an alkenyl radical, a cycloalkyl radical, an aryl group, an aralkyl

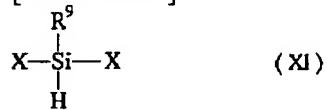
radical, an alkylamino radical, an alkyl silyl radical, or an alkoxy group independently among an upper type, respectively, R7 is the radical of bivalence, and X is a halogen atom.)

[Claim 6] The manufacture approach of the silicon content copolymerization polymer characterized by the mixture containing the disilyl compound expressed with the ORGANO PORIHARO silane and general formula (IX) which are expressed with said general formula (VIII) in the manufacture approach of a silicon content table polymerization polymer according to claim 5 being what manufactures a silicon content copolymerization polymer according to claim 2 including the ORGANO PORIHARO silane further expressed with the following general formula (X) and/or a general formula (XI).

[Formula 12]



[Formula 13]



(R8 and R9 are an alkyl group, an alkenyl radical, a cycloalkyl radical, an aryl group, an aralkyl radical, an alkylamino radical, an alkyl silyl radical, or an alkoxy group independently among an upper type, respectively, and X is a halogen atom.)

[Claim 7] The mixture containing the disilyl compound expressed with the ORGANO PORIHARO silane and general formula (IX) which are expressed with said general formula (VIII) in the first stage story of a reaction The manufacture approach of the silicon content copolymerization polymer according to claim 3 characterized by making it react with the diamine expressed with the following general formula (XII), making the water which the solvent suitable subsequently was made to distribute react, carrying out a reaction with ammonia after that and making an unreacted halo silane react completely.

NH2-R10-NH2 (XII)

(R10 is the aromatic series radical of bivalence among a formula.)

[Claim 8] The manufacture approach of the silicon content copolymerization polymer according to claim 6 or 7 characterized by the mixture containing the disilyl compound expressed with the ORGANO PORIHARO silane and general formula (IX) which are expressed with said general formula (VIII) containing the ORGANO PORIHARO silane further expressed with said general formula (X) and/or general formula (XI).

[Claim 9] The film which consists of a silicon content copolymerization polymer according to claim 1 to 4.

[Translation done.]

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention is excellent in a new silicon content copolymerization polymer and its manufacture approach, and the thermal resistance further formed of this new silicon content copolymerization polymer at transparency, and its specific inductive capacity is low and it relates to the film excellent in flexibility and a mechanical strength. Moreover, the new silicon content copolymerization polymer of this invention can be broadly used for the part to which the thermal resistance and the transparency, and also the low dielectric constant of heat-resistant coating besides a film, the dielectric layer for plasma display panels (PDP), a semi-conductor interlayer insulation film, etc. are demanded. Since especially the silicon content copolymerization polymer of this invention is excellent in the transparency after hardening in atmospheric air and can make specific inductive capacity low, it is suitable as a dielectric layer ingredient for PDP.

[0002]

[Description of the Prior Art] Since silicone was compounded by Rocho and others about the thermally stable polymer, aromatic series and heterocycle macromolecules, such as a fluorine system ingredient, polyimide vendor ZORU, aromatic polyamide, and polyimide, have been studied. Furthermore, a heat-resistant demand increases with development of space and the aeronautical-navigation field, progress of research is seen in aromatic series and a heterocycle system macromolecule, and amelioration of aromatic polyimide is advanced in recent years. Moreover, the organic metal polymer which a principal chain becomes from metallic elements, such as Si, Ti, and B, O, N, etc. like a PORIBORO siloxane and the poly CHITANO siloxane in addition to this is studied.

[0003] Heat-resistant improvement is desired in a heatproof and insulating material fields various in recent years. For example, the rotating-machine fields, such as a generator, a high-pressure alternating current motor, an industrial direct-current motor, and an electric-car motor, are mentioned as one of the applications of a heatproof and an insulating material. The thing belonging to these high-pressure rotating machines has an inclination to large-capacity-izing and high-pressure-izing, and a miniaturization and high thermal resistance are required. And the heat-resistant demand characteristics in these fields have some which exceed 400 degrees C among atmospheric air. Furthermore, in addition to this, insulation, flexibility, a mechanical property, etc. are needed with thermal resistance.

[0004] As other applications of a heatproof and an insulating material, it is Pre-Metal-Dielectric corresponding to an insulation of a display device or dielectric layers, such as a dielectric layer for PDP, a semi-conductor interlayer insulation film, and 600-degree-C low-temperature process. Various things including the heat-resistant film used for semiconductor device ingredients, such as a Layer (PMD) ingredient, the front plate of a display, and various applications or a heat-resistant sheet, heat-resistant coating, a plastic optical fiber, a wire covering, etc. are mentioned. In addition to the thermal resistance like the above, flexibility, and a mechanical property, in these applications, optical properties, such as transparency and dielectric characteristics, and electrical characteristics are also needed. Furthermore, use is tried also as an ingredient of fields, such as an advanced-features product with ceramic ingredients, such as ceramic composite, ceramic-metal-to-metal adhesive, aluminum₂O₃, and MgO, and an advanced-features product by the blend with other polymers. However, the heat-resistant temperature put in practical use about the organic polymer among current thermally stable polymers is 300 degrees C or less.

[0005] On the other hand, since the thermal stability of a polymer is high, organic metal polymers, such as silicone resin and PORIBORO siloxane resin, have the high thermal resistance of 500 degrees C or more. however, a line with little organic macromolecule to branching of the molecular structure -- to being structure, the organic metal polymer has the network structure with much branching, the huge three-dimensional network was formed of the dehydrogenation condensation reaction of OH radicals, or oxidation reaction, as a result, it was lacking in flexibility,

and the use application was restricted.

[0006]

[Problem(s) to be Solved by the Invention] Then, in order to solve the fault of such an organic metal polymer, the block copolymerization silazane which compound-ization with organic resin and an inorganic material etc. is tried, for example, consists of an inorganic polysilazane part and an organic polysilazane part is proposed (for example, JP,2-175726,A). however, a such compound-ized polymer also has an inadequate mechanical property -- etc. -- it was not what there is a problem and can still be satisfied.

[0007] In view of such a situation, this invention person developed the high thermal-resistance silicon content copolymerization polymer which consists of a silazane copolymer including a specific repeat unit (JP,8-231727,A and JP,9-188765,A). This silicon content copolymerization polymer has the thermal resistance of 400 degrees C or more, and it is the organic metal polymer excellent in a mechanical property and flexibility, and that manufacture is also easy a polymer and it has the description it is featureless to it being transparent by hardening in nitrogen. Therefore, although it is an ingredient useful for a high heatproof optical application, when it hardens in atmospheric air, the obtained hardening polymer is colored and there is a problem that it is difficult to obtain the film with good transparency. For this reason, in order to obtain a transparent hardening polymer, it needs to be hardened in nitrogen, and it is generated also when the application for an application from process cost and a mass-production side is difficult. For this reason, in order to raise usefulness more, it is the further technical problem to develop what cannot spoil thermal resistance but can discover hardening processing in nitrogen and the transparency more than comparable also by hardening processing in atmospheric air. Moreover, while excelling in such thermal resistance, a mechanical property, flexibility, and transparency, the polymer with low specific inductive capacity is also called for.

[0008] Therefore, the purpose of this invention solved the above-mentioned technical problem, i.e., it has the thermal resistance of 400 degrees C or more, excels in a mechanical strength and flexibility, and has high transparency by colorlessness also by hardening in atmospheric air, and it is to offer a silicon content copolymerization polymer also with low specific inductive capacity, and its manufacture approach. Moreover, other purposes of this invention are excellent in the thermal resistance and transparency which were formed of the silicon content copolymerization polymer which has the property which was excellent the account of a top, a mechanical strength, and flexibility, and are to offer a film also with low specific inductive capacity.

[0009]

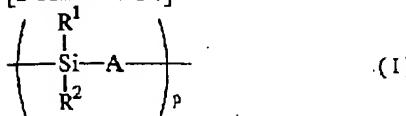
[Means for Solving the Problem] As a result of this invention person's repeating examination wholeheartedly, it became clear that the coloring in the case of hardening processing in the atmospheric air of the polymer proposed by said JP,8-231727,A and JP,9-188765,A has a large part resulting from the diamine used for manufacture of a polymer. Namely, although diamine is used as a modifier at the time of polymer manufacture and silazane association is introduced into a polymer in the silicon content copolymerization polymer developed previously It was realized that it became possible to suppress the coloring in the case of hardening in atmospheric air by coming to guess that it is because the meeting condition after hardening of the diamine component introduced at this time has influenced coloring of a hardening polymer, and controlling the condition of diamine. For that, it is possible to reduce the amount of denaturation of diamine as a concrete means, and to lower reaction temperature. However, if it is a component required for linear-izing of a heavy polymer, and control of polymerization degree and silicon content decreases this, polymerization degree will fall, and diamine can consider that physical properties, such as the thermal resistance of a polymer and reinforcement, fall. In order to compensate this point, it becomes clear that aim at installation of Si-O association which is comparatively newly easy to carry out a polymerization to a linear, and the transparent film is obtained by reduction of the amount of diamine denaturation and the fall of reaction temperature also in 400-degree C hardening among atmospheric air with installation of this Si-O association, and that the improvement in transparency and decline in specific inductive capacity are moreover also seen, and this invention is reached. it becomes that whose industrial usefulness of a heat-resistant silicon content copolymerization polymer was markedly alike, and improved by this invention.

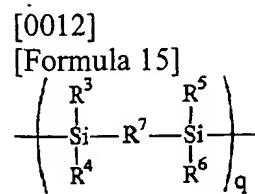
[0010] This invention consists of the following configurations.

(1) The silicon content copolymerization polymer which number average molecular weight is 500-1,000,000, and is characterized by including the structural unit expressed with the following general formula (I) and (II) at least.

[0011]

[Formula 14]





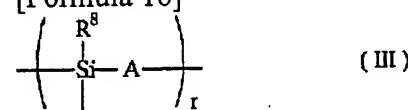
(II)

[0013] R1, R2, R3, R4, R5, and R6 are an alkyl group, an alkenyl radical, a cycloalkyl radical, an aryl group, an aralkyl radical, an alkylamino radical, an alkyl silyl radical, or an alkoxy group independently among [upper type, respectively, R7 is the radical of bivalence, and A is NH or O. Moreover, a structural unit (I) and (II) are random, each mole ratio p and q is $q/(p+q) = 0.01-0.99$, and the rate of Si-O association in a polymer and Si-N association is $Si-O/(Si-N+Si-O) = 0.01-0.99$.]

[0014] (2) a silicon content copolymerization polymer -- further -- the following general formula (III) and a silicon content copolymerization polymer given in (IV) above-mentioned [which is characterized by including a kind of structural unit at least] (1).

[0015]

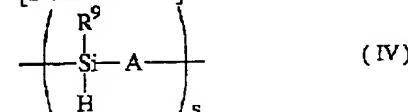
[Formula 16]



(III)

[0016]

[Formula 17]



(IV)

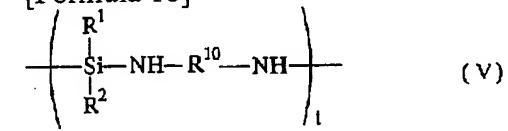
[0017] R8 and R9 are an alkyl group, an alkenyl radical, a cycloalkyl radical, an aryl group, an aralkyl radical, an alkylamino radical, an alkyl silyl radical, or an alkoxy group independently among [upper type, respectively, and A is NH or O. Moreover, the structural unit in a polymer (I) - (IV) are random, and each mole ratio p, q, r, and s takes the following relation.

 $q/(p+q+r+s) = 0.01-0.99$

[0018] (3) a silicon content copolymerization polymer -- further -- the following general formula (V) and (VI) -- and (VII) a silicon content copolymerization polymer the above (1) characterized by including a kind of structural unit at least, or given in (2).

[0019]

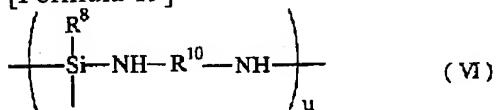
[Formula 18]



(V)

[0020]

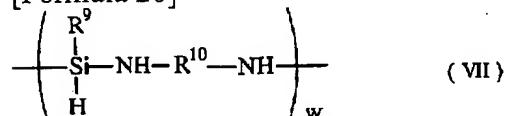
[Formula 19]



(VI)

[0021]

[Formula 20]



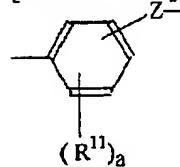
(VII)

[0022] R1, R2, R8, and R9 are an alkyl group, an alkenyl radical, a cycloalkyl radical, an aryl group, an aralkyl radical, an alkylamino radical, an alkyl silyl radical, or an alkoxy group independently among [upper type, respectively, and R10 is the aromatic series radical of bivalence. Moreover, a structural unit (V) - (VII) are random.]

[0023] (4) A silicon content copolymerization polymer given in either of above-mentioned (1) - (3) characterized by the aromatic series radical of said bivalence being a radical expressed with an aralkylene group, a naphthylene radical, or the following general formula (A).

[0024]

[Formula 21]

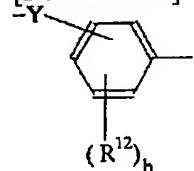


(A)

[0025] R11 is a radical which a halogen atom or a low-grade alkyl group, and a couple directly the integer of 0-4, and Z, or is expressed with the following general formula (B) among [upper type.

[0026]

[Formula 22]



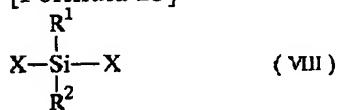
(B)

[0027] (or [that R12 couples directly the integer of 0-4, and Y for a halogen atom or a low-grade alkyl group, and b among a formula] -- or it is the radical of bivalence.)]

[0028] (5) The manufacture approach of the silicon content copolymerization polymer the above-mentioned (1) publication characterized by making the mixture which contains at least the disilyl compound expressed with the ORGANO PORIHARO silane expressed with the following general formula (VIII), and the following general formula (IX) react with the water which the suitable solvent was made to distribute, carrying out a reaction with ammonia after that and making an unreacted halo silane react completely.

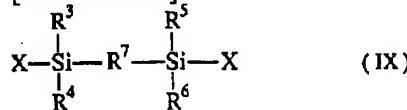
[0029]

[Formula 23]



[0030]

[Formula 24]

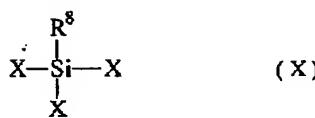


[0031] (R1, R2, R3, R4, R5, and R6 are an alkyl group, an alkenyl radical, a cycloalkyl radical, an aryl group, an aralkyl radical, an alkylamino radical, an alkyl silyl radical, or an alkoxy group independently among an upper type, respectively, R7 is the radical of bivalence, and X is a halogen atom.)

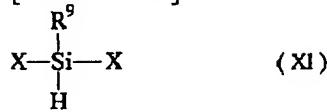
[0032] (6) The manufacture approach of the silicon content copolymerization polymer characterized by for the mixture containing the disilyl compound expressed with the ORGANO PORIHARO silane and the general formula (IX) which are expressed with said general formula (VIII) to the above (5) in the manufacture approach of the silicon content table polymerization polymer a publication to be what manufactures the silicon content copolymerization polymer of the above-mentioned (2) publication including the ORGANO PORIHARO silane further expressed with the following general formula (X) and/or a general formula (XI).

[0033]

[Formula 25]



[0034]
[Formula 26]



[0035] (R8 and R9 are an alkyl group, an alkenyl radical, a cycloalkyl radical, an aryl group, an aralkyl radical, an alkylamino radical, an alkyl silyl radical, or an alkoxy group independently among an upper type, respectively, and X is a halogen atom.)

[0036] (7) The mixture containing the disilyl compound expressed with the ORGANO PORIHARO silane and general formula (IX) which are expressed with said general formula (VIII) in the first stage story of a reaction The manufacture approach of the silicon content copolymerization polymer the above-mentioned (3) publication characterized by making it react with the diamine expressed with the following general formula (XII), making the water which the solvent suitable subsequently was made to distribute react, carrying out a reaction with ammonia after that and making an unreacted halo silane react completely.

NH₂-R10-NH₂ (XII)

(R10 is the aromatic series radical of bivalence among a formula.)

[0037] (8) The manufacture approach of a silicon content copolymerization polymer the above (6) characterized by the mixture containing the disilyl compound expressed with the ORGANO PORIHARO silane and general formula (IX) which are expressed with said general formula (VIII) containing the ORGANO PORIHARO silane further expressed with said general formula (X) and/or general formula (XI), or given in (7).

[0038] (9) The above (1) Film which becomes either of - (4) from the silicon content copolymerization polymer of a publication.

[0039] In addition, according to this invention, the manufacture approach of the further following silicon content copolymerization polymer is offered as a desirable mode.

(i) The manufacture approach of the silicon content copolymerization polymer the aforementioned (4) publication that the radical of the bivalence of R7 is characterized by being the radical expressed with an aralkylene group, a naphthylene radical, or said general formula (A) among a general formula (IX).

(ii) The manufacture approach of the silicon content copolymerization polymer the aforementioned (5) publication that the aromatic series radical of the bivalence of R10 is characterized by being the radical expressed with an aralkylene group, a naphthylene radical, or said general formula (A) among a general formula (XII).

[0040] Furthermore, according to this invention, the manufacture approach of the following polymer and a polymer is offered as a more desirable mode.

** The aforementioned silicon content copolymerization polymer R1-R6, and whose R8 and R9 are a methyl group or a phenyl group independently in the structural unit of said general formula (I) - (VII), respectively.

** The aforementioned silicon content copolymerization polymer R7 and whose R10 are arylene radicals in said general formula (II) and the structural unit of (V) - (VII).

[0041] ** The manufacture approach of the aforementioned silicon content copolymerization polymer that R1-R6, and R8 and R9 are a methyl group or a phenyl group independently in the structural unit of said general formula (VIII) - (XI), respectively.

** Said general formula (IX) And (XII) the manufacture approach of the aforementioned silicon content copolymerization polymer that R7 and R10 are arylene radicals in a compound.

** The manufacture approach of the aforementioned silicon content copolymerization polymer that X is Cl atom in the compound of said general formula (VIII) - (XI).

[0042] Moreover, although the coating film containing the rebound ace court film and films, such as a film simple substance, are mentioned as a use application of the silicon content copolymerization polymer of this invention, the use mode like a less or equal can be illustrated besides it.

(a) It is PDP vacuum encapsulant to the object for PDP tooth-back plates and the dielectric layer for front plates which consist of the aforementioned silicon content copolymerization polymer, and the charge list of PDP rib (septum) material.

- (b) The interlayer insulation film of the semiconductor device which consists of the aforementioned silicon content copolymerization polymer.
- (c) Coating which consists of the aforementioned silicon content copolymerization polymer.
- (d) The front plotting board of the display device which consists of the aforementioned silicon content copolymerization polymer.
- (e) The PMD ingredient corresponding to 600-degree-C low-temperature process which consists of the aforementioned silicon content copolymerization polymer.
- (f) The optical fiber which consists of the aforementioned silicon content copolymerization polymer.
- (g) Adhesives which consist of the aforementioned silicon content copolymerization polymer.
- (h) The wire covering which consists of the aforementioned silicon content copolymerization polymer.
- (i) Composite which consists of an aforementioned silicon content copolymerization polymer and the aforementioned ceramics.

[0043]

[Embodiment of the Invention] Hereafter, this invention is explained in detail concretely. The silicon content copolymerization polymer of this invention has said general formula (I) and (II), and the structural unit expressed with (I) - (IV), or (I) - (VII) by the case, and is characterized by number average molecular weight being in the range of 500-1,000,000. The copolymerization polymer of this invention from having said general formula (II) and the structural unit further expressed with (V) - (VII) by the case Binding energy goes up by C=C association, and the linearization of a polymer advances by installation of two functional groups (it is [R7 and] R10 by the case), and, of course, by Si association, since oxidation stability is high. Since it has the thermal resistance of 400 degrees C or more, and it has flexibility, and has said general formula (III) by installation of three functional groups and moreover has the structural unit of (VI) by the case upwards, a mechanical strength will become high. In said general formula (I) and (III) the -NH-radical list in (V) - (VII), furthermore, by the general formula (IV) and (VII) existence of inner Si-H. Since hardening can progress with heating, and what has strong reinforcement can be further obtained with thermal resistance and it has Si-O association, a hardening polymer is colorlessness, when it excels in transparency by colorlessness and moreover hardens in atmospheric air, and can form the hardening polymer which has the outstanding transparency. The general formula (IV) which has Si-H, and (VII) control of hardening when the structural unit of a structural unit and the general formula (I) which has -NH-, (III), (IV) and (V) - (VII) exists, in case it is heat hardening can be performed easily.

[0044] the structural unit as which the silicon content copolymerization polymer of this invention is expressed in (I) - (IV) or (I), (II) and (V) - (VII), or (I) - (VII) by the general formula (I) and (II), and the case as mentioned above -- having -- especially -- (I) -- or (III) -- and the description is in the point of having Si-O association in the structure expressed with (IV). In addition, the joint sequence of each configuration unit of a general formula (I) - (VII) is random, and the ratios p, q, r, and s of each component, or p, q, r, s, t, u and w can take the following range.

[0045] $p/(p+q+r+s) = 0.01-0.99$ -- desirable -- 0.1-0, and $5q/(p+q+r+s) = 0.01-0.99$ -- desirable -- 0.2-0.75r/(p+q+r+s) = 0-0.99 -- desirable -- 0.1-0.5s/(p+q+r+s) = 0-0.99 -- desirable -- 0.01-0.2 [0046] or $p/ (p+q+r+s+t+u+w) = 0.01-0.99$ -- desirable -- 0.1-0.5q/(p+q+r+s+t+u+w) = 0.01-0.99 -- desirable -- 0.1-0.75 (t+u+w)/(p+q+r+s+t+u+w) = 0-0.99 -- desirable -- 0.01-0.5 (second+w)/(p+q+r+s+t+u+w) = 0-0.99 -- desirable -- 0.01-0.2 (r+u)/(p+q+r+s+t+u+w) = 0-0.99 -- desirable -- 0.1-0.75 [0047] Furthermore, although the rate of Si-O association in a polymer and Si-N association is Si-O/(Si-N+Si-O) = 0.01-0.99, they are 0.1-0.95 preferably.

[0048] Next, the manufacture approach of the silicon content copolymerization polymer of this invention is explained. The mixture containing the disilyl compound expressed with the ORGANO PORIHARO silane as which the manufacture approach of this invention is expressed in said general formula (VIII), and said general formula (IX), Or the mixture with which the ORGANO PORIHARO silane further expressed with said general formula (X) and/or general formula (XI) to this if needed was contained It is characterized by making it react with the water which the suitable solvent was made to distribute after making it react with the diamine first expressed with said general formula (XII), when required, carrying out a reaction with ammonia after that and making an unreacted halo silane react completely.

[0049] In the ORGANO PORIHARO silane expressed with the general formula (VIII) used as a start raw material in case the silicon content copolymerization polymer in this invention is manufactured, (X), or (XI) Although R1, R2, R8, and R9 are chosen from an alkyl group, an alkenyl radical, a cycloalkyl radical, an aryl group, an aralkyl radical, an alkylamino radical, an alkyl silyl radical, or an alkoxy group usually, a carbon number -- 1-7 -- desirable -- 1-5 -- further -- desirable -- the alkyl group of 1-2 -- a carbon number -- the alkenyl radical of 2-7, and a carbon number -- the cycloalkyl radical of 5-7, and an aryl group -- general -- as X -- usually -- a fluorine, chlorine, a bromine, and an iodine atom -- a chlorine atom is used preferably. As an aryl group, a phenyl group, a tolyl group, a xylyl group, a

KUMENIRU radical, benzyl, a phenethyl radical, alpha-methylbenzyl radical, a benzhydryl group, a trityl radical, a styryl radical, a cinnamyl radical, a biphenyl radical, a naphthyl group, etc. can be used. As an alkyl silyl radical (monochrome, JI, tree substitution product), an alkylamino radical (monochrome, G substitution product), and an alkoxy group, what has 1-7 carbon atoms is usually used. In addition, R1 and R2 may be the same, and they may differ, respectively. As a compound expressed with said general formula (VIII), diphenyl dichlorosilane is desirable, as a compound expressed with said general formula (X), phenyl trichlorosilane is desirable and methyl dichlorosilane is desirable as a compound expressed with said general formula (XI).

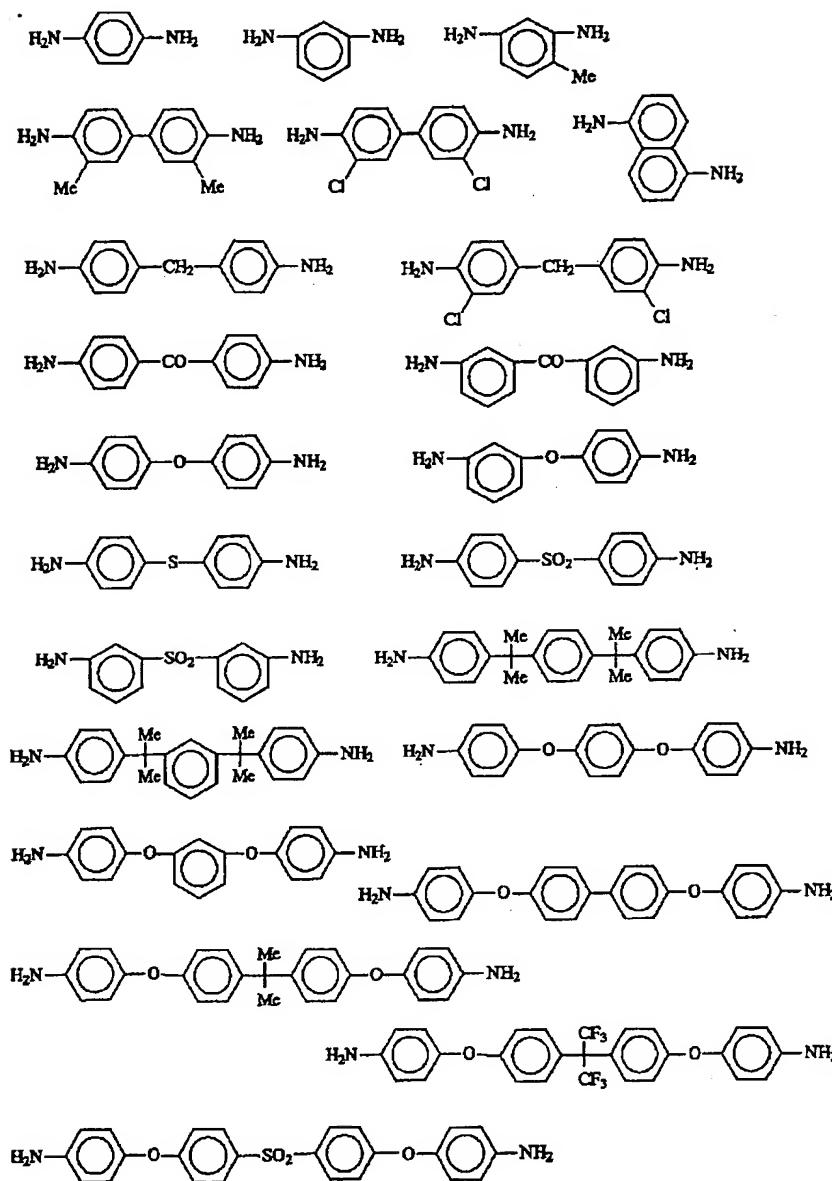
[0050] R3-R6 of a disilyl compound which are expressed with the general formula (IX) used as a start raw material on the other hand in case the silicon content copolymerization polymer in this invention is manufactured are chosen from an alkyl group, an alkenyl radical, a cycloalkyl radical, an aryl group, an aralkyl radical, an alkylamino radical, an alkyl silyl radical, or an alkoxy group like R1, R2, R8, and R9 of a general formula (VIII), (X), and (XI), and X is a halogen atom. The thing in R1, R2, R8, and R9 in the ORGANO PORIHARO silane as which the example of these radicals or an atom is expressed in a general formula (VIII), (X), and (XI), and the same thing are mentioned. Moreover, as a radical of the bivalence of R7, the aromatic series radical of bivalence, such as a radical expressed with an aralkylene group, a naphthylene radical, or said general formula (A), is desirable. As R7, an alkylene group, an alkenylene group, a cyclo alkylene group, an arylene radical, an alkyl imino group, or an alkyl silylene radical is mentioned, and, specifically, an arylene radical is desirable.

[0051] Moreover, as an arylene radical, a phenylene group, a tolylene radical, a xylylene radical, a benzylidene radical, a FENE dust DIN radical, alpha-methyl benzylidene radical, the Singh Nami Liden radical, a naphthylene radical, etc. can be mentioned. As a compound expressed with a general formula (IX), 1 and 4-bis(dimethyl chlorosilyl) benzene etc. is specifically desirable.

[0052] Mixture with the above-mentioned ORGANO PORIHARO silanes and a disilyl compound is made to react in this invention approach if needed with the diamine first expressed with general formula (XII) NH₂-R10-NH₂. As for R10 in a general formula (XII), it is desirable that it is the aromatic series radical of bivalence and is the radical expressed with an aralkylene group, a naphthylene radical, or said general formula (A). The example of diamine shown in Table 1 by the general formula (XII) is given. An arylene radical is desirable, although aromatic series radicals of various bivalence, such as arylene radicals, such as a phenylene group, and a biphenylene radical, are mentioned as shown in this example as R10. In addition, although the example of diamine is shown in Table 1, it does not pass for these to have shown as an only desirable example, and the diamine expressed with the general formula (XII) of this invention is not restricted to a thing given in Table 1. Especially in such diamines, the Para-phenylenediamine (p-PDA), meta-phenylenediamine (m-PDA), 4, and 4'-diphenyl diamino ether (oxy-dianiline, ODA) is desirable.

[0053]

[Table 1]



[0054] Moreover, as a reaction solvent, any of the independent or mixture of a Lewis base and a nonresponsive solvent may be used. in this case -- as a Lewis base -- for example, tertiary amine (a trimethylamine --) Torr alkylamines, such as dimethyl ethylamine, diethyl monomethylamine, and triethylamine, A pyridine, picoline, dimethyl ARININ and these derivatives, and the secondary amine that have the radical of steric hindrance nature phosphoretted hydrogen, SUCHIPIN, arsines, these derivatives (for example, trimethyl phosphoretted hydrogen --), etc. Dimethyl ethyl phosphoretted hydrogen, methyl diethyl phosphoretted hydrogen, triethyl phosphoretted hydrogen, a trimethyl arsine, trimethyl SUCHIPIN, a trimethylamine, triethylamine, etc. can be mentioned. Especially, a basic small base (for example, a pyridine, picoline, trimethyl phosphoretted hydrogen, dimethyl ethyl phosphoretted hydrogen, methyl diethyl phosphoretted hydrogen, triethyl phosphoretted hydrogen) is more desirable than ammonia at a low-boiling point, and especially a pyridine and picoline are desirable from handling and economy.

[0055] Moreover, as a nonresponsive solvent, ether, such as the halogenated hydrocarbon; aliphatic series ether, such as hydrocarbon solvent; halogenation methane of aliphatic hydrocarbon, alicyclic hydrocarbon, and aromatic hydrocarbon, halogenation ethane, and halogenation benzene, and alicyclic ether, can be used. One desirable also in these A methylene chloride, chloroform, a carbon tetrachloride, Bromoform, an ethylene chloride, an ethylidene chloride, trichloroethane, Halogenated hydrocarbon, such as tetrachloroethane, ethyl ether, isopropyl ether, Ethyl butyl ether, butyl ether, 1, 2-dioxy ethane, Ether, such as dioxane, dimethyl dioxane, a tetrahydrofuran, and tetrahydropyran, A pentane, a hexane, an isohexane, a methyl pentane, a heptane, an iso heptane, They are hydrocarbons, such as an octane, an isoctane, a cyclopentane, methylcyclopentane, a cyclohexane, a methylcyclohexane, benzene, toluene, a xylene, ethylbenzene, a N-methyl-2-pyrrolidone, and a jig lime. The dichloromethane from points, such as safety, a

xylene, and a N-methyl-2-pyrrolidone are desirable among these solvents. Moreover, a pyridine / dichloromethane mixed solvent is also desirable.

[0056] By this invention approach, the disilyl compound first expressed with said general formula (VIII), and the ORGANO PORIHARO silane expressed with a general formula (IX) and (X) if required and a general formula (XI) is mixed in an organic solvent. Then, although the diamine first expressed with said general formula (XII) will be made to react if required in a mole ratio, that what is necessary is just within the limits of 1:99-99:1, the mixed rates of the ORGANO PORIHARO silane and a disilyl compound are 90:10-10:90 preferably, and are 80:20-40:60 still more preferably. Moreover, although what is necessary is for the range of the operating rate of both the above-mentioned halo silicon compound and diamine just to be 100:0-10:90 in a mole ratio, they are 100:0-25:75 preferably and are 100:0-40:60 still more preferably. Moreover, as an amount of amine denaturation by diamine, it is desirable to carry out to 0-50-mol% of the theoretical reacting weight of both the halo silicon compound. Although the concentration of the halo silicon compound in a solvent can be chosen as arbitration, it is good to consider as 1 - 25% of the weight of the range. as for temperature, the system of reaction serves as a liquid -- it may learn the range (typically -40 degrees C - 300 degrees C), and a gap is sufficient. Moreover, although a pressure is generally under ordinary pressure - pressurization, the bottom of nitrogen pressurization is good.

[0057] After carrying out the reaction of said halo silicon compound and diamine, the water which the proper solvent was made to distribute is made to add and react to a halo silicon compound, without performing a reaction with diamine, and Si-O association is made to generate. The thing same as a solvent which distributes water at this time as the solvent in a reaction with the above-mentioned diamine can be used. Especially a pyridine and picoline are desirable from handling and economy. Moreover, in a reaction with water, the grouting velocity of the water to the system of reaction has big effect on generation of a polymer. When grouting velocity is quick, generation of a polymer may not fully be performed. As grouting velocity of water, less than [0.1molH₂O/min] is desirable. Furthermore, a role also with reaction temperature important for generation of a polymer is played. -40 degrees C - 20 degrees C of temperature of a hydro RISHISU reaction are usually -20 degrees C - 5 degrees C more preferably. When reaction temperature is high, generation of a polymer may not fully be performed.

[0058] After a reaction with water is completed, ammonia is added, an aminolysis reaction is carried out and a halo silane is made to react completely. Conditions, such as a reaction solvent in this case and reaction temperature, are the same as a case with the diamine of the preceding paragraph. The addition of ammonia is decided by the amount of the halogen atom which remains without reacting. That is, although the amount of theory required for the aminolysis of a halo silicon compound is calculable from the amount of the water measurement of the added diamine, since ammonia may become superfluous, it is used in an amount usually more superfluous than the amount of theory. Although a pressure is generally under [from under ordinary pressure] pressurization, the bottom of nitrogen pressurization is desirable. Although HCl generates in this reaction, this can make a salt from bases, such as triethylamine or ammonia, and can be separated with the quality of the specified substance. Thus, a copolymerization polymer will be obtained, if the manufactured copolymerization polymer, byproduct ammonium chloride, or amine salt is carried out a ** exception and a solvent is removed under reduced pressure of this filtrate.

[0059] According to the approach of this invention, it has said general formula (I) and (II), and the structural unit expressed with (I) - (IV) or (I), (II) and (V) - (VII), or (I) - (VII) by the case, and the new silicon content copolymerization polymer which has number average molecular weight in the range of 500-1,000,000 is obtained easily. In addition, this polymer is meltable to common organic solvents, such as the hydrocarbon solvent of aliphatic hydrocarbon, alicyclic hydrocarbon, and aromatic hydrocarbon, halogenated hydrocarbon, ether, alcohol, ester, and a ketone.

[0060] Moreover, the copolymerization polymer obtained by this invention approach can be used as high thermal resistance and a Plastic solid with the outstanding mechanical strength. For example, a film simple substance can be obtained by obtaining a coating film, if a copolymer is applied on a glass substrate and calcinated from 0.05 at 250 to 550 degrees C in atmospheric air or the inert atmosphere of nitrogen, an argon, etc. for 2.0 hours, and exfoliating this from a glass substrate. These films are films which it has the mechanical property and flexibility which bore and were excellent in the elevated temperature, and **** is mostly excellent in transparency no perfect coloring, and have low specific inductive capacity.

[0061]

[Example] Hereafter, although an example explains this invention to a detail further, the technical range of this invention is not limited by these.

[0062] After desiccation nitrogen permuted the inside of the reaction container installed in the thermostat, phenyl trichlorosilane (PhSCl₃) 47gr (0.222 mols), diphenyl dichlorosilane (Ph₂SiCl₂) 56gr (0.222 mols), methyl

dichlorosilane (MeSiCl₂) 3.8gr (0.033 mols) and 1, and the thing in which 4-bis(dimethyl chlorosilyl) benzene 50gr (0.19 mols) was dissolved were supplied to xylene 1000ml. Next, when whenever [reaction container internal temperature] is set as -5 degrees C and the solution reached predetermined temperature, DDE(diamino diphenyl ether) 3.56gr (0.0178 mols) is supplied, and it stirred for 1 hour and was made to react. Then, in order to crush hydrochloride generation of DDE generated at the reaction, triethylamine 40gr was dissolved in xylene 500ml, and it poured into the reaction container. Stirring was continued after impregnation for 1 hour. The solution made to dissolve water (H₂O) 13.0gr (0.7222 mols) in pyridine 1000ml was poured into the reaction container at the rate of about 30 ml/min, maintaining the temperature in a reaction container at -5 degrees C. At this time, the reaction of a halo silane and water occurred with impregnation, and whenever [container internal temperature] went up to -2 degrees C. After impregnation of water and a pyridine mixed solution was completed, stirring was continued for 1 hour. Then, the making unreacted chlorosilane react completely purpose, ammonia was poured in for 10 minutes at the rate of 2 NL/min, and was stirred. Generation of white precipitate of an ammonium chloride was checked with addition of ammonia. After reaction termination, after blowing desiccation nitrogen and removing unreacted ammonia, pressure filtration was carried out under nitrogen-gas-atmosphere mind, and about 2300ml of filtrate was obtained. When the solvent permutation of this filtrate was carried out under reduced pressure, the transparent high viscosity resin which 105gr colored yellow slightly was obtained.

[0063] The number average molecular weight of the obtained resin was 2,100. Moreover, the result of IR-spectrum analysis, To absorption; 1060-1100cm⁻¹ based on Si-Ph absorption; 1020-820cm⁻¹ based on Si-O absorption; 1140cm⁻¹ based on Si-H to absorption; 2160cm⁻¹ based on a N-H radical to wave number 3350cm⁻¹ Si-H And absorption based on Si-N-Si; it was checked by 3140, 2980, and 1270m⁻¹ the absorption 810 based on C-H, and 780cm⁻¹ that the absorption based on C-H of the benzene ring is shown.

[0064] the place which furthermore analyzed 1 H-NMR (proton nuclear magnetic resonance absorption) spectrum of this polymer -- delta 7.2 -- ppm (br, C₆H₆) and delta 4.8 -- ppm (br, SiH) and delta 1.4 -- ppm (br, NH) and delta Absorption of 0.3 ppm (br, SiCH₃) was checked, and it was judged as that from which the quality of the specified substance was obtained.

[0065] Next, the physical properties of this polymer were investigated. Applied this polymer to the thickness of 50 microns on the glass substrate, it was made to harden on 500 degrees C and the conditions of 1 hour among 400 degrees C, 1 hour, and nitrogen among atmospheric air, respectively, and measurement of permeability and specific inductive capacity was carried out. Consequently, there were also no generating and exfoliation of a crack, it is 97% and, as for each permeability, heat-resistant temperature also showed the permeability and thermal resistance of 550 degrees C or more which were very excellent.

[0066] The silicon content copolymerization polymer was compounded according to the approach indicated by the example 1 of example JP,9-188765,A of a comparison, spreading postcure was carried out on the glass substrate, and it examined like the example 1. Consequently, permeability was 93%, although 400 degrees C was hardened in atmospheric air for 1 hour and 500 degrees C of permeability were hardened among nitrogen 40% for 1 hour. Moreover, the specific inductive capacity of the film hardened 400 degrees C among 400 degrees C, 1-hour hardening, and nitrogen among atmospheric air for 1 hour was 3.08 and 3.0.

[0067] From the result of an example 1 and the example of a comparison, the remarkable improvement of the permeability in the inside of atmospheric air was accepted by installation of Si-O association. The polymer obtained in the example 1 has the thermal resistance exceeding polyimide, and the permeability more than polymethylmethacrylate (PMMA). what has such thermal resistance and light transmission nature in existing resin -- not existing -- therefore, high heatproof optical application equal width (dielectric layer of a plasma display etc.) -- large application is possible. Moreover, compared with the polyimide film conventionally used widely as a heat-resistant film, it excels in thermal resistance and light transmittance, and since specific inductive capacity is also low, the use as an ingredient which replaces a polyimide film is possible.

[0068] Moreover, as a result of measuring the specific inductive capacity of the hardening film of the polymer of the example 1 respectively hardened at 400 degrees C, 500 degrees C, and 600 degrees C among nitrogen as an example of electric physical properties for 1 hour, they were 2.72, 2.71, 2.54, and a low dielectric constant, respectively. The application as a next-generation semi-conductor interlayer insulation film and a dielectric application for PDP is possible for the polymer of such thermal resistance and specific inductive capacity, and transparency to the example 1.

[0069] The silicon content heatproof polymer was obtained like the example 1 except having changed into the conditions which show the addition of two to example 4 water, the class of use diamine, and the amount of denaturation in Table 2. Furthermore, transmissometry of the obtained polymer was performed like the example 1. A result is shown in Table 3.

[0070]
[Table 2]

	水の添加量 (mol%)	ジアミン種類	ジアミン変性量 (mol%)	数平均分子量
実施例1	93.5	DDE	5.0	2,100
実施例2	93.5	P・PDA	5.0	2,050
実施例3	93.5	—	0	2,200
実施例4	70.0	DDE	25.0	1,800
比較例	0	P・PDA	50.0	850

[0071]
[Table 3]

	透過率 (%)	
	大気中 400°C、1時間硬化	窒素中 500°C、1時間硬化
実施例1	97	97
実施例2	97	97
実施例3	97	97
実施例4	95	96
比較例	40	93

[0072] In addition, in order to compare the specific inductive capacity of the polymer of examples 1-4 and the example of a comparison, 400 degrees C of each polymers were hardened in atmospheric air and nitrogen for 1 hour, and specific inductive capacity was measured. A result is shown in Table 4.

[0073]
[Table 4]

	比誘電率	
	大気中 400°C、1時間硬化	窒素中 400°C、1時間硬化
実施例1	2.75	2.72
実施例2	2.74	2.73
実施例3	2.73	2.74
実施例4	2.83	2.81
比較例	3.08	3.00

[0074]
[Effect of the Invention] As stated above, since it has the high thermal resistance of about 550 degrees C, about 97% of Takamitsu permeability, and low specific inductive capacity, and a mechanical strength also needs to be strong, the polymer which shows flexibility can be obtained and it is not necessary to set up special conditions, such as nitrogen-gas-atmosphere mind, also by hardening in atmospheric air, industrial use for various applications is attained by low cost by this invention.

[Translation done.]